

**PCT**WORLD INTELLECTUAL PROPERTY ORGANIZATION  
International Bureau

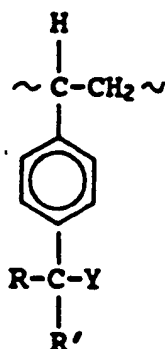
## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

<b>(51) International Patent Classification <sup>5</sup> :</b> C08F 2/54, 210/10 C09D 125/04, G03F 7/027 C08L 23/22	<b>A1</b>	<b>(11) International Publication Number:</b> WO 92/11295 <b>(43) International Publication Date:</b> 9 July 1992 (09.07.92)
<b>(21) International Application Number:</b> PCT/US91/09653 <b>(22) International Filing Date:</b> 19 December 1991 (19.12.91)  <b>(30) Priority data:</b> 633,645                      20 December 1990 (20.12.90)    US  <b>(71) Applicant:</b> EXXON CHEMICAL PATENTS INC. [US/ US]; 1900 East Linden Avenue, Linden, NJ 07036 (US).  <b>(72) Inventors:</b> AUDETT, Jay, Douglas ; N38 W 6812 Wilson Drive, Cedarburg, WI 53012 (US). DIAS, Anthony, Jay ; 1411 Quiet Green Ct., Houston, TX 77062 (US). POW- ERS, Kenneth, William ; 145 Robbins Avenue, Berkeley Heights, NJ 07922 (US). WANG, Hsien, Chang ; 14 Whittier Street, Edison, NJ 08820 (US).		<b>(74) Agent:</b> SCHNEIDER, John, E.; Exxon Chemical Com- pany, P.O. Box 710, 1900 East Linden Avenue, Linden, NJ 07036 (US).  <b>(81) Designated States:</b> AT (European patent), BE (European patent), CH (European patent), DE (European patent), DK (European patent), ES (European patent), FR (Eu- ropean patent), GB (European patent), GR (European patent), IT (European patent), JP, LU (European pa- tent), MC (European patent), NL (European patent), SE (European patent).  <b>Published --</b> <i>With international search report. Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i>
<b>(54) Title:</b> UV/EB CURABLE BUTYL COPOLYMERS FOR COATING APPLICATIONS  <b>(57) Abstract</b>  A radiation reactive functionalized polymer comprising an isoolefin having about 4 to about 7 carbon atoms and a para-alkylstyrene, wherein a radiation reactive functional group is attached to the para-alkyl group of the para-alkylstyrene. The polymer has a substantially homogeneous compositional distribution and may be utilized in lithographic and coating applications. Also disclosed are radiation curable pressure sensitive adhesives comprising the functionalized polymer and a tackifier.		

## CLAIMS:

- 1 1. A radiation-curable coating composition, comprising:
  - 2 (a) a radiation-crosslinkable functionalized  
3 polymer of an isocolefin of 4 to 7 carbon  
4 atoms and para-alkylstyrene comonomer,  
5 said functionalized polymer having  
6 substantially homogeneous compositional  
7 distribution wherein the comonomer is  
8 functionalized with a radiation-reactive  
9 group at the para-alkyl group of the para-  
10 alkylstyrene; and  
11 (b) a tackifier resin.
- 1 2. The coating composition of claim 1, wherein said  
2 functionalized polymer has a number average  
3 molecular weight of at least about 5000.
- 1 3. The coating composition of claim 2, wherein said  
2 functionalized polymer has a number average  
3 molecular weight of between about 5000 and 500,000.
- 1 4. The coating composition of claim 1, wherein said  
2 functionalized polymer has a ratio of weight average  
3 molecular weight to number average molecular weight  
4 of less than about 6.
- 1 5. The coating composition of claim 1, wherein at least  
2 about 95 weight percent of said functionalized  
3 polymer has a para-alkylstyrene content within about  
4 10 weight percent of the average para-alkylstyrene  
5 content for said functionalized polymer.
- 1 6. The coating composition of claim 1, wherein said  
2 isocolefin comprises isobutylene and said para-  
3 alkylstyrene comprises para-methylstyrene.
- 1 7. The coating composition of claim 6, wherein said  
2 functionalized polymer has a number average  
3 molecular weight greater than about 5000.
- 1 8. The coating composition of claim 7, wherein said  
2 functionalized polymer has a ratio of weight average  
3 molecular weight to number average molecular weight  
4 of less than about 6.

- 1 9. The coating composition of claim 1, wherein said  
 2 isoolefin and said para-alkylstyrene are present in  
 3 said functionalized polymer in amounts such that  
 4 said isoolefin comprises from about 10 to 99.5  
 5 percent by weight of said polymer and said para-  
 6 alkylstyrene comprises from about 0.5 to 90 percent  
 7 by weight of said functionalized polymer.
- 1 10. The coating composition of claim 9, wherein said  
 2 isoolefin comprises isobutylene and said para-  
 3 alkylstyrene comprises para-methylstyrene.
- 1 11. The coating composition of claim 1, wherein said  
 2 copolymer includes said para-alkylstyrene as

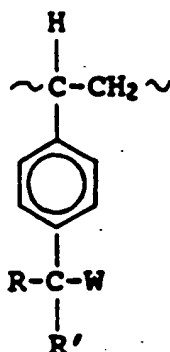


- 15 wherein R and R' are independently selected from the  
 16 group consisting of hydrogen, alkyl, and primary and  
 17 secondary alkyl halide radicals and Y comprises a  
 18 mixture of hydrogen and radiation-reactive  
 19 functionality.
- 1 12. The coating composition of claim 11, wherein R and  
 2 R' are independently selected from the group  
 3 consisting of hydrogen, C<sub>1</sub> to C<sub>3</sub> alkyl and C<sub>1</sub> to C<sub>3</sub>  
 4 primary and secondary alkyl halide.

- 1 13. The coating composition of claim 11, wherein said  
2 radiation-reactive functionality comprises a  
3 nucleophilically substituted photoinitiator  
4 selected from the group consisting of benzophenone,  
5 4-chlorobenzophenone, 4-hydroxybenzophenone, benzo-  
6 quinone, naphthaquinone, anthraquinone, 2-  
7 chloroanthraquinone, benzylidene acetophenone,  
8 acetophenone, propiophenone, cyclopropyl phenyl  
9 ketone, benzaldehyde,  $\beta$ -naphthylphenyl ketone,  $\beta$ -  
10 naphthaldehyde,  $\beta$ -acetoneaphthone, 2,3-pentanedione,  
11 benzil, fluorenone, benzanthrone, Michler's ketone,  
12 bis(parahydroxybenzylidene)acetone, benzoin, deoxy-  
13 benzoin, and chlorodeoxybenzoin.
- 1 14. The coating composition of claim 11, wherein said  
2 radiation-reactive functionality comprises  
3 nucleophilically substituted thioxanthone.
- 1 15. The coating composition of claim 11, wherein said  
2 radiation-reactive functionality comprises a  
3 nucleophilically substituted photoinitiator selected  
4 from N,N-disubstituted dithiocarbamic acid and  
5 esters and salts thereof.
- 1 16. The coating composition of claim 11, wherein said  
2 radiation-reactive functionality comprises a  
3 nucleophilically substituted photoinitiator selected  
4 from tung oil acid and esters and salts thereof.
- 1 17. The coating composition of claim 11, wherein said  
2 radiation-reactive functionality comprises  
3 photoinitiators selected from the group consisting  
4 of benzoic acid, cinnamic acid, m-nitrocinnamic  
5 acid, p-chlorocinnamic acid, p-methoxycinnamic acid,  
6 chalcone acrylic acid, p-phenylenebis(acrylic acid)  
7 p-azidobenzoic acid, p-sulfonazidebenzoic acid,  $\alpha$ -  
8 cyanocinnamic acid, cinnamylideneacetic acid,  
9 cinnamylidenemalonic acid,  $\alpha$ -cyanocinnamylidene-  
10 acetic acid,  $\beta$ -(1)-naphthylacrylic acid,  $\beta$ -(2)-  
11 furfurylacrylic acid,  $\alpha$ -cyano- $\beta$ -(2)-thienylacrylic  
12 acid,  $\beta$ -(1) naphthylacrylic acid,  $\beta$ -(9)-  
13 anthrylacrylic acid and esters and salts thereof.

- 1 18. The coating composition of claim 1, wherein said  
2 polymer comprises from about 5 to about 95 parts by  
3 weight of the composition and said tackifier  
4 comprises from about 5 to about 95 parts by weight  
5 of the composition, wherein parts by weight of the  
6 polymer and the tackifier total 100.
- 1 19. The coating composition of claim 1, wherein said  
2 polymer comprises from about 30 to about 70 parts by  
3 weight of the composition and said tackifier  
4 comprises from about 30 to about 70 parts by weight  
5 of the composition, wherein parts by weight of the  
6 polymer and the tackifier total 100.
- 1 20. The coating composition of claim 1, wherein said  
2 copolymer comprises from about 10 to about 99.5  
3 weight percent of said isoolefins and from about 0.5  
4 to about 90 weight percent of said para-  
5 alkylstyrene.

- 1 21. The coating composition of claim 20, wherein said  
 2 para-alkylstyrene is included in said polymer as:



15 wherein R and R' are independently selected from  
 16 hydrogen, alkyl and primary and secondary alkyl  
 17 halide, and W is selected from hydrogen, X, Y and Z,  
 18 wherein X is chlorine or bromine, Y is a radiation-  
 19 reactive functional group, and Z is a non-radiation-  
 20 reactive functional group containing oxygen, sulfur,  
 21 silicon, nitrogen, carbon or metal selected from  
 22 sodium potassium, lithium and magnesium, wherein  
 23 said para-alkylstyrene wherein W is hydrogen  
 24 comprises from about 0.5 to about 99.5 percent by  
 25 weight of said polymer, said para-alkylstyrene,  
 26 wherein W is Y comprises from about 0.5 to about 55  
 27 percent by weight of said polymer, said para-  
 28 alkylstyrene, wherein W is X comprises from 0 to 55  
 29 percent by weight of said polymer, and said para-  
 30 alkylstyrene wherein W is Z comprises from 0 to 55  
 31 percent by weight of said polymer, said polymer  
 32 having a number average molecular weight above about  
 33 5000.

- 1 22. The coating composition of claim 21, wherein said  
2 para-alkylstyrene wherein W is hydrogen comprises  
3 from about 1 to about 20 percent by weight of said  
4 polymer, said para-alkylstyrene wherein W is Y  
5 comprises from about 0.5 to about 20 percent by  
6 weight of said polymer, said para-alkylstyrene  
7 wherein W is X comprises less than about 20 percent  
8 by weight of said polymer, and said para-  
9 alkylstyrene wherein W is Z comprises from 0 to  
10 about 20 percent by weight of said polymer, said  
11 polymer having a number average molecular weight  
12 from about 5000 to about 500,000 and a ratio of  
13 weight average molecular weight to average molecular  
14 weight less than about 6.
- 1 23. The coating composition of claim 21, wherein said  
2 para-alkylstyrene wherein W is hydrogen comprises  
3 from about 2 to about 10 percent by weight of said  
4 polymer, said para-alkylstyrene wherein W is Y  
5 comprises from about 0.5 to about 15 percent by  
6 weight of said polymer, said para-alkylstyrene  
7 wherein W is X comprises less than about 15 percent  
8 by weight of said polymer, and said para-  
9 alkylstyrene wherein W is Z comprises from 0 to  
10 about 15 percent by weight of said polymer, said  
11 polymer having a number average molecular weight  
12 from about 5000 to about 500,000 and a ratio of  
13 weight average molecular weight to average molecular  
14 weight less than about 6.
- 1 24. The coating composition of claim 21, wherein said  
2 para-alkylstyrene wherein W is Y comprises from  
3 about 1 to about 7 percent by weight of said  
4 polymer, and said para-alkylstyrene wherein W is X  
5 comprises less than about 1 percent by weight of  
6 said polymer, said polymer having a number average  
7 molecular weight from about 5000 to about 500,000  
8 and a ratio of weight average molecular weight to  
9 average molecular weight less than about 6.

- 1 25. The coating composition of claim 21, wherein said  
2 polymer is essentially free of said para-  
3 alkylstyrene wherein W is X.
- 1 26. The coating composition of claim 22, wherein said  
2 polymer comprises less than about 1 percent by  
3 weight of said para-alkylstyrene wherein W is X.
- 1 27. A process for producing a radiation-crosslinkable  
2 functionalized polymer, comprising reacting:  
3 (a) in a nucleophilic substitution reaction, a  
4 halogenated polymer comprising an  
5 isocolefin of 4 to 7 carbon atoms and a  
6 para-alkylstyrene comonomer, said polymer  
7 having a substantially homogeneous  
8 compositional distribution and essentially  
9 free of halogen atoms in the polymer  
10 backbone and also in the aromatic rings,  
11 said halogen atoms being substantially  
12 alkylhalide attached to the pendant para-  
13 alkylstyrene and primarily  
14 alkylmonohalide; with  
15 (b) a nucleophilic reagent comprising a  
16 radiation reactive nucleophile.
- 1 28. The process of claim 27, further comprising the  
2 preliminary steps of reacting said isocolefin and  
3 said para-alkylstyrene in a polymerization reactor  
4 under polymerization conditions in the presence of a  
5 diluent and a Lewis Acid catalyst, maintaining said  
6 polymerization reactor substantially free of  
7 impurities which can complex with said catalyst or  
8 copolymerize with said isocolefin or said para-  
9 alkylstyrene to obtain a precursor copolymer, and  
10 contacting said precursor copolymer with a halogen  
11 in the presence of a free radical initiator to  
12 obtain said halogenated polymer.
- 1 29. The process of claim 27, wherein said isocolefin  
2 comprises isobutene and said para-alkylstyrene  
3 comprises para-methylstyrene.
- 1 30. The process of claim 27, wherein said halogen  
2 comprises bromine.



1 31. The process of claim 27, wherein said radiation  
2 reactive nucleophile comprises a nucleophilically  
3 substituted photoinitiator selected from the group  
4 consisting of benzophenone, 4-chlorobenzophenone, 4-  
5 hydroxybenzophenone, benzoquinone, naphthaquinone,  
6 anthraquinone, 2-chloroanthraquinone, benzylidene  
7 aceto-phenone, acetophenone, propiophenone,  
8 cyclopropyl phenyl ketone, benzaldehyde,  $\beta$ -  
9 naphthylphenyl ketone,  $\beta$ -naphthaldehyde,  $\beta$ -  
10 acetoneaphthone, 2,3-pentanedione, benzil,  
11 fluorenone, benzanthrone, Michler's ketone,  
12 bis(parahydroxybenzylidene)acetone, benzoin, deoxy-  
13 benzoin, and chlorodeoxybenzoin.

1 32. The process of claim 27, wherein said radiation  
2 reactive nucleophile comprises nucleophilically  
3 substituted thioxanthone photoinitiator.

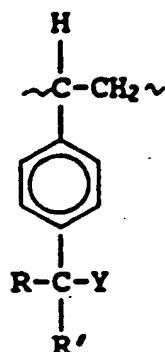
1 33. The process of claim 27, wherein said radiation  
2 reactive nucleophile comprises a nucleophilically  
3 substituted photoinitiator selected from N,N-  
4 disubstituted dithiocarbamic acid and esters and  
5 salts thereof.

1 34. The process of claim 27, wherein said radiation  
2 reactive nucleophile comprises a nucleophilically  
3 substituted photoinitiator selected from tung oil  
4 acid and esters and salts thereof.

1 35. The process of claim 27, wherein said radiation  
2 reactive nucleophile comprises a nucleophilically  
3 substituted photoinitiator selected from benzoic  
4 acid, cinnamic acid, m-nitrocinnamic acid, p-  
5 chlorocinnamic acid, p-methoxycinnamic acid,  
6 chalcone acrylic acid, p-phenylenebis(acrylic acid)  
7 p-azidobenzoic acid, p-sulfonazidebenzoic acid,  $\alpha$ -  
8 cyanocinnamic acid, cinnamylideneacetic acid,  
9 cinnamylidenemalonic acid,  $\alpha$ -cyanocinnamylidene-  
10 acetic acid,  $\beta$ -(1)-naphthylacrylic acid,  $\beta$ -(2)-  
11 furfuryl-acrylic acid,  $\alpha$ -cyano- $\beta$ -(2)-thienylacrylic  
12 acid,  $\beta$ -(1) naphthylacrylic acid,  $\beta$ -(9)-  
13 anthrylacrylic acid and esters and salts thereof.

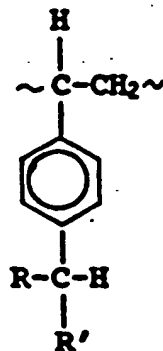
- 1 36. A coated article having adhered to at least a  
2 portion of a surface thereof, the radiation curable  
3 coating composition of claim 1.
- 1 37. The article of claim 36, wherein said surface is  
2 coated with a hot melt of said coating composition.
- 1 38. The article of claim 36, wherein said coating  
2 composition is crosslinked by exposure to  
3 electromagnetic radiation.
- 1 39. A method for making a coated article, comprising the  
2 steps of:  
3 (a) coating at least a portion of a surface of  
4 the article with the coating composition  
5 of claim 1;  
6 (b) exposing said surface to electromagnetic  
7 radiation to crosslink the coating  
8 composition.
- 1 40. The method of claim 39, wherein said surface is  
2 coated with a hot melt of said coating composition.
- 1 41. The method of claim 39, wherein said crosslinking  
2 radiation comprises electron beam or gamma  
3 radiation.

- 1 42. A radiation curable copolymer, comprising an  
2 isoolefin of 4 to 7 carbon atoms randomly  
3 polymerized with a para-alkylstyrene comonomer,  
4 wherein the para-alkylstyrene is included as

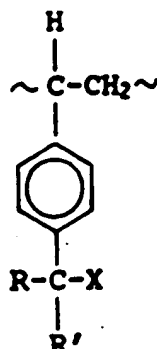


17 wherein R and R' are independently selected from the  
18 group consisting of hydrogen, alkyl and primary and  
19 secondary alkyl halides, and Y is a radiation-  
20 reactive functional group added by nucleophilic  
21 substitution of a photoinitiator compound selected  
22 from one or more of aromatic aldehydes and ketones,  
23 alkoxy and acyl-substituted aromatics, hetero  
24 aromatics, fused ring polycyclic aromatics,  
25 dithiocarbamates, unsaturated fatty acids, aromatic  
26 carboxylic acids, nitroaromatics, dye compounds,  
27 azides, diazonium salts and combinations thereof.

- 1 43. The copolymer of claim 42, wherein said para-  
2 alkylstyrene is further included as

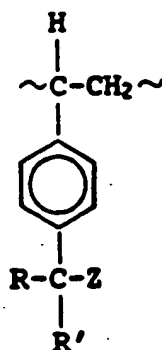


- 1 44. The copolymer of claim 42, wherein said para-  
2 alkylstyrene is further included as



16 wherein X is halogen.

- 1 45. The copolymer of claim 42, wherein said para-  
2 alkylstyrene is further included as




16 wherein Z is a non-radiation-reactive functional  
17 group containing oxygen, sulfur, silicon, nitrogen,  
18 carbon, phosphorus or a metal selected from sodium,  
19 potassium, lithium, and magnesium.

- 1 46. The copolymer of claim 42, wherein said  
2 photoinitiator comprises a nucleophilically  
3 substituted photoinitiator selected from the group  
4 consisting of benzophenone, 4-chlorobenzophenone, 4-  
5 hydroxybenzophenone, benzoquinone, naphthaquinone,  
6 anthraquinone, 2-chloroanthraquinone, benzylidene  
7 aceto-phenone, acetophenone, propiophenone,  
8 cyclopropyl phenyl ketone, benzaldehyde,  $\beta$ -  
9 naphthylphenyl ketone,  $\beta$ -naphthaldehyde,  $\beta$ -  
10 acetonaphthone, 2,3-pentanedione, benzil,  
11 fluorenone, benzanthrone, Michler's ketone,  
12 bis(parahydroxybenzylidene)acetone, benzoin,  
13 deoxybenzoin, and chlorodeoxybenzoin.
- 1 47. The copolymer of claim 42, wherein said  
2 photoinitiator comprises nucleophilically  
3 substituted thioxanthone.
- 1 48. The copolymer of claim 42, wherein said  
2 photoinitiator comprises a nucleophilically  
3 substituted photoinitiator selected from N,N-  
4 disubstituted dithiocarbamic acid and esters and  
5 salts thereof.
- 1 49. The copolymer of claim 42, wherein said  
2 photoinitiator comprises a nucleophilically  
3 substituted photoinitiator selected from tung oil  
4 acid and esters and salts thereof.
- 1 50. The copolymer of claim 42, wherein said  
2 photoinitiator comprises photoinitiators selected  
3 from the group consisting of benzoic acid, cinnamic  
4 acid, m-nitrocinnamic acid, p-chlorocinnamic acid,  
5 p-methoxycinnamic acid, chalcone acrylic acid, p-  
6 phenylenebis(acrylic acid) p-azidobenzoic acid, p-  
7 sulfonazidebenzoic acid,  $\alpha$ -cyanocinnamic acid,  
8 cinnamylideneacetic acid, cinnamylidenemalononic acid,  
9  $\alpha$ -cyanocinnamylideneacetic acid,  $\beta$ -(1)-naphthyl-  
10 acrylic acid,  $\beta$ -(2)-furfurylacrylic acid,  $\alpha$ -cyano- $\beta$ -  
11 (2)-thienylacrylic acid,  $\beta$ -(1) naphthylacrylic acid,  
12  $\beta$ -(9)-anthrylacrylic acid and esters and salts  
13 thereof.

- 1 51. A coated article, comprising an article having  
2 adhered to at least a portion of a surface thereof,  
3 the radiation-curable copolymer of claim 42.
- 1 52. The article of claim 51, wherein said surface is  
2 coated with a hot melt of the copolymer.
- 1 53. The article of claim 51, wherein said copolymer is  
2 crosslinked by exposure to electromagnetic  
3 radiation.
- 1 54. A method for making a coated article, comprising the  
2 steps of:  
3 (a) coating at least a portion of a surface of  
4 the article with the copolymer of claim  
5 42;  
6 (b) exposing said surface to electromagnetic  
7 radiation to crosslink the copolymer.
- 1 55. The method of claim 54, wherein said coating  
2 comprises a hot melt or solvent application.
- 1 56. The method of claim 54, wherein said crosslinking  
2 radiation comprises ultraviolet, electron beam or  
3 gamma radiation.

## INTERNATIONAL SEARCH REPORT

International Application No PCT/US 91/09653

<b>I. CLASSIFICATION OF SUBJECT MATTER</b> (If several classification symbols apply, indicate all) <sup>6</sup>		
According to International Patent Classification (IPC) or to both National Classification and IPC		
IPC5: C 08 F 2/54, 210/10, C 09 D 125/04, G 03 F 7/027 C 08 L 23/22		
<b>II. FIELDS SEARCHED</b>		
Minimum Documentation Searched <sup>7</sup>		
Classification System	Classification Symbols	
IPC5	C 08 F; C 08 L; C 09 D; G 03 F	
Documentation Searched other than Minimum Documentation to the extent that such Documents are included in Fields Searched <sup>8</sup>		
<b>III. DOCUMENTS CONSIDERED TO BE RELEVANT<sup>9</sup></b>		
Category <sup>10</sup>	Citation of Document <sup>11</sup> with indication, where appropriate, of the relevant passages <sup>12</sup>	Relevant to Claim No. <sup>13</sup>
E, Y	WO, A1, 9119761 (EXXON CHEMICAL PATENTS INC) 26 December 1991, see page 8, line 16 - line 24; page 9, line 4 - line 16; page 10, line 1 - line 20; page 17, line 10 - line 14 page 18, line 9 - line 20; page 20, line 1 - line 7  --	1-12
Y	EP, A2, 0344021 (EXXON CHEMICAL PATENTS INC) 29 November 1989, see column 2, line 38 - line 61; column 3, line 31 - line 34; column 4, line 51 - line 60; column 5, line 31 - line 36  --	1-12
<p><sup>10</sup> Special categories of cited documents: <sup>13</sup></p> <p><sup>A</sup> document defining the general state of the art which is not considered to be of particular relevance</p> <p><sup>E</sup> earlier document but published on or after the international filing date</p> <p><sup>L</sup> document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p><sup>O</sup> document referring to an oral disclosure, use, exhibition or other means</p> <p><sup>P</sup> document published prior to the international filing date but later than the priority date claimed</p> <p><sup>T</sup> later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</p> <p><sup>X</sup> document of particular relevance, the claimed invention cannot be considered novel or cannot be considered to involve an inventive step</p> <p><sup>Y</sup> document of particular relevance, the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art</p> <p><sup>A</sup> document member of the same patent family</p>		
<b>IV. CERTIFICATION</b>		
Date of the Actual Completion of the International Search	Date of Mailing of this International Search Report	
13th April 1992	29. 04. 92	
International Searching Authority	Signature of Authorized Officer	
EUROPEAN PATENT OFFICE	Nicole De Blé 	

III. DOCUMENTS CONSIDERED TO BE RELEVANT (CONTINUED FROM THE SECOND SHEET)		
Category *	Citation of Document, with indication, where appropriate, of the relevant passages	Relevant to Claim No
Y	US, A, 4556464 (DAVID J. ST. CLAIR) 3 December 1985, see page 3, line 8 - line 13; page 3, line 47 - line 58; page 7, line 5 - line 7; page 7, line 21 - line 34; page 8, line 23 - line 31; page 9, line 1 - line 15, line 25 - line 34; page 12, line 40 - line 45; page 15, line 62  --	1-12
A,P	US, A, 5013793 (HSIEN C. WANG ET AL) 7 May 1991, see column 4, line 6 - line 10; column 4, line 30 - line 42; column 4, line 57 - line 60; column 5, line 13 - line 36; column 7, line 19  --	1-8, 11
A,P	WO, A1, 9104992 (EXXON CHEMICAL PATENTS INC) 18 April 1991, see page 2, line 2 - line 28; page 3, line 17 - line 27; page 4, line 7 - line 9; page 5, line 1 - line 14; page 8, line 24  --	1-8,10- 12
A	US, A, 4489020 (DANIEL J. DIBIASI) 18 December 1984, see column 2, line 13; claims 1-2  --	1
A	US, A, 4779657 (SEUNG T. CHEUNG ET AL) 25 October 1988, see column 2, line 21 - line 40; column 3, line 25 - line 29  --	1
A	US, A, 4716183 (JOSE P. GAMARRA ET AL) 29 December 1987, see column 3, line 35 - line 40; column 3, line 54; claim 1  --	1



III. DOCUMENTS CONSIDERED TO BE RELEVANT (CONTINUED FROM THE SECOND SHEET)		
Category *	Citation of Document, with indication, where appropriate, of the relevant passages	Relevant to Claim No
A	US, A, 3948863 (KENNETH W. POWERS) 6 April 1976, see column 3, line 15; claim 1  -----  -----	1

**ANNEX TO THE INTERNATIONAL SEARCH REPORT  
ON INTERNATIONAL PATENT APPLICATION NO. PCT/US 91/09653**

SA 55792

This annex lists the patent family members relating to the patent documents cited in the above-mentioned international search report. The members are as contained in the European Patent Office EDP file on 28/02/92. The European Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
WO-A1- 9119761	26/12/91	NONE	
EP-A2- 0344021	29/11/89	AU-B- 612845 AU-D- 3521989 JP-A- 2150408	18/07/91 30/11/89 08/06/90
US-A- 4556464	03/12/85	NONE	
US-A- 5013793	07/05/91	WO-A- 92/01746	06/02/92
WO-A1- 9104992	18/04/91	AU-D- 4658389 AU-D- 4842390 WO-A- 91/04995	28/04/91 28/04/91 18/04/91
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